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Waxy bitumen stranding in southern Australia: A geochemical study of multiple oil families and their likely origins

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ABSTRACT

Reports of bitumen stranding on the ocean beaches of southern Australia date back to the early days of European settlement. Previous investigations have shown that this 'coastal bitumen' comprises three categories of stranded petroleum: waxy bitumen, asphaltite and oil slicks. All three varieties are physically and chemically distinct from each other, and bear no geochemical resemblance to any indigenous Australian crude oil. This study focuses on the most common variety, waxy bitumen, which accounted for 90% of the strandings on six South Australian beaches repeatedly surveyed during 1991–1992. Geochemical analysis of 96 individual specimens collected from these survey sites and other beaches in South Australia and western Victoria has shown them to be variously weathered high-wax crude oils of paraffinic to aromatic-intermediate bulk composition. Elemental, isotopic and biomarker differences allow their assignment to at least five oil families with inferred source facies that range from deep freshwater lacustrine through paludal and deltaic to euxinic marine, possibly deposited within different sedimentary basins. Family 1, 2 and 3 waxy bitumens all contain biomarkers derived from the freshwater alga *Botryococcus* sp. and tropical angiosperms (notably dipterocarps). Similar biomarker assemblages are unknown in Australian sedimentary basins but are common in Cenozoic crude oils and source rocks throughout western Indonesia. Family 4 waxy bitumens lack these biomarkers, but do contain dinosterane and 24-*n*-propylcholestane, indicative of a marine source affinity, while the carbon isotopic signatures and high pristane/phytane (Pr/Ph) ratios of Family 5 waxy bitumens are consistent with their origin from coal-rich source rocks deposited in fluvial to deltaic sedimentary successions. The majority of these waxy bitumens represent an oceanic influx of non-indigenous, Southeast Asian crude oils carried into the waters of southern Australia by the Leeuwin Current. Although they are likely to originate from natural seepage within the Indonesian Archipelago, it is unknown whether the parent oils emanate from submarine seeps or from inland seepages which are then carried to the sea by rivers. The common practice of tanker cleaning operations in the Java and Banda seas may augment the supply of natural bitumen to the beaches of Australia.

1. Introduction

A recent comprehensive review by Edwards et al. (2016) outlined the history and geographic distribution of bitumen stranding on ocean beaches along the southern coastline of Australia. While these 'coastal bitumens' can readily be distinguished from the occasional spillages of crude and refined petroleum from passing maritime traffic (Padley et al., 1993), they are by no means uniform in either appearance or composition. Historical records reveal that there are two main types of this 'ocean wanderer' bitumen: hard asphalt-like material ('asphaltum', now known as asphaltite) and softer waxy bitumen (tar balls).

Arguably the most distinctive variety, asphaltites are a single family of heavy (4–18 °API gravity) aromatic-asphaltic crudes with very high sulfur contents (3–4%). Their biomarker and isotopic signatures indicate an origin from marine shale deposited during a mid-Cretaceous oceanic anoxic event (Edwards et al., 1998; Totterdell et al., 2008). They are most likely a product of low-

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intensity seepage from tar mats exposed by the incision of submarine canyons into the continental slope along Australia's southern margin (Hall et al., 2014). While asphaltite appears to have been the main variety referred to in reports from the earlier part of the twentieth century, waxy bitumen is now the most common form of Australian coastal bitumen, accounting for 90% of annual strandings in South Australia and western Victoria during the early 1990s (Padley, 1995; Edwards et al., 2016).

1.1. Historical continuity of waxy bitumen stranding

Tar balls have been stranding on the beaches of southern Australia for more than 100 years, long before the advent of ocean-going tankers and local shipping powered by fuel oil (Sprigg and Woolley, 1963). Moreover, the range of chemical compositions evident in these stranded bitumens has remained relatively constant for at least 65 years (Edwards et al., 2016). They are typically found on sandy west-facing ocean beaches close to the high-water mark. However, there are exceptions.

Among the samples analysed in the present study are several examples of highly weathered 'fossil waxy bitumen'. These were found within a pumice-rich sand layer above a prominent calcrete horizon located close to the base of a coastal sand dune at the top of the present-day beach, as well as on the inland side of this dune in Beachport Conservation Park (Edwards et al., 2016, their Fig. 4). Fossil waxy bitumen samples were also found in blowouts between and behind barrier dunes within Canunda National Park and as far south as the Admella Dunes (Fig. 1). These Quaternary barrier shoreline deposits, which are preserved along the Coorong Coastal Plain (Bourman et al., 2016) and extend as far south as Portland in Victoria, have been extensively dated using thermoluminescence (Huntley et al., 1993) and amino acid racemisation (Murray-Wallace et al., 2001) techniques. Since the dune is located above present sea level and within the youngest Robe Range (I) coastal sand barrier system, equivalent in age to the Younghusband Peninsula that fronts the Coorong Lagoon, the pumice-rich layer and its contained bitumen may be up to 7 ka in age (Bourman et al., 2016). Geochemically, these fossil bitumen samples appear almost identical to the main family of waxy bitumens now being washed up on southern Australian beaches, thereby negating the possibility of an anthropogenic origin.

1.2. Previous geochemical studies

The first systematic attempt to characterise the geochemistry of these waxy bitumens was that of McKirdy and Horvath (1976). Using API gravity, US Bureau of Mines (USBM) correlation indices, bulk composition, elemental and isotopic analyses, gas chromatography and trace metal contents, they distinguished two types of waxy bitumen (highly paraffinic to paraffinic, and paraffinic-naphthenic), and inferred that both were derived from early mature lacustrine Cretaceous source rocks, suggesting that they may have emanated from seafloor seepage in the western Otway Basin. However, the analytical techniques employed in this initial study did not permit unambiguous determination of their source affinity.

In 1983 the South Australian Department of Mines and Energy (SADME) conducted a major survey of stranded bitumen along the South Australian coastline, involving the collection of new samples for more sophisticated analysis, including by gas chromatography–mass spectrometry (GC–MS), in order to promote petroleum exploration within the adjacent offshore Otway and Duntroon basins. Interpretation of the resulting data by McKirdy (1984a, 1984b) led to the recognition of four genetically distinct families (Families 1, 2, 3 and 4; the first three comprising waxy bitumens and the last asphaltite) based on their sulfur content, the carbon isotopic composition of their C_{12+} saturated and aromatic hydrocarbons, and various alkane biomarker ratios; criteria later augmented by other elemental data (C, H and N) and bulk hydrogen and sulfur isotopic signatures (McKirdy, 1985a).

Subsequent publications (McKirdy, 1985b; McKirdy et al., 1986, 1988) adopted the aforementioned threefold classification for the southern Australian waxy bitumens and continued to maintain that they were likely to have originated from lacustrine source beds within the Early Cretaceous rift-fill of the Otway Basin. However, failure to identify their distinctive biomarker assemblage (botryococcane, bicadinanes, oleanane and 4-methylsteranes) in any organic-rich facies of this basin (Padley, 1995) necessitated a wider research area. The fact that this association of biomarkers has so far been found only in Cenozoic lacustrine source rocks and crude oils from Sumatra and Java (Seifert and Moldowan, 1981; Williams et al., 1985; Longley et al., 1990; Schiefelbein and Cameron, 1997; Hwang et al., 2002), now suggests that the waxy bitumens represent flotsam which originates from oil seeps in the Indonesian Archipelago and is carried southward into Australian waters by long-distance ocean currents (Currie et al., 1992; McKirdy et al., 1994; Padley, 1995; McGowran et al., 1997; Edwards et al., 2016). That said, there is no agreement on the number of different oil families represented (Table 1), and the locations of their parent seeps are yet to be identified.

1.3. Present investigation

This paper presents the results of geochemical analyses undertaken by Padley (1995) on a suite of 96 archival and more recently stranded waxy bitumens recovered from ocean beaches between Cape Otway, Victoria and the southern Eyre Peninsula, South Australia (Fig. 1). Most samples (n = 53) were collected during 1991 and 1992. When combined with elemental, isotopic, biomarker and bulk compositional information from other sources (McKirdy, 1984a, 1984b, 1985a; Tewkesbury, 1989; Watson, 1990), the resulting compilation provided the basis for a revised genetic classification of southern Australian waxy coastal bitumens, and a clearer understanding of their weathering characteristics, source affinity and likely origins. The latter was achieved, first by comparison of our updated classification with those of bitumens found in the Northern Territory (Summons et al., 1993) and Western Australia (Alexander et al., 1994); and then with the various oil families identified in the western Indonesian Archipelago (Robinson, 1987; ten Haven and Schiefelbein, 1995; Schiefelbein and Cameron, 1997; Hwang et al., 2002).

2. Materials and methods

2.1. Sample suite

The bitumen samples examined in this study are listed in Supplementary Table S1, along with their date of collection and, where available, API gravity and sulfur content. The locations of their respective stranding sites are shown in Fig. 1. Each sub-sample taken for analysis came from the interior of the tar ball in order to minimise the effect of weathering on the resulting analytical data. To document these effects in one of the larger specimens (sample 28), three sub-samples were removed from its interior, intermediate and exterior zones and analysed separately.

2.2. Sample fractionation

A weighed aliquot of pure coastal bitumen was dissolved in a minimum amount of dichloromethane and flooded with petroleum ether (30 mL/g). The solution was then refluxed for 2 h in the

Fig. 1. Map showing the location of waxy bitumen stranding sites in South Australia and western Victoria.

presence of activated copper turnings to remove elemental sulfur. The insoluble asphaltenes were separated from the soluble maltenes (asphaltene-free oil) by filtration through pre-extracted filter paper (Whatman Grade 42). The asphaltenes were washed with petroleum ether in a Soxhlet apparatus and then recovered by dissolving in dichloromethane. Solvent was removed from the asphaltene and maltene fractions under vacuum in a Büchi rotary evaporator. Each fraction, dissolved in a small volume of solvent, was transferred by Pasteur pipette to a pre-weighed glass vial. Final removal of the solvent was achieved under a stream of nitrogen.

Maltenes were separated into hydrocarbons (saturated and aromatic) and polar compounds (resins) by standard liquid chromatography on a column of activated silica over alumina

Table 1
Previous geochemical studies of Australian waxy coastal bitumen.

Location	No. of samples	No. of oil families	Data source
South Australia & Northern Territory	11	2	1
South Australia & western Victoria	27	3	2
South Australia & western Victoria	96	5	3
Northern Territory	22	2	4
Western Australia	70	8	5

1. McKirdy and Horvath (1976); 2. McKirdy et al. (1984a, 1984b, 1985b, 1986, 1994); 3. Padley (1995); 4. Summons et al. (1993); 5. Alexander et al. (1994).

(sample: adsorbent ratio = 1:100). The saturated hydrocarbon fraction was eluted with petroleum ether (80 mL); mono-, di- and tri-aromatic hydrocarbon fraction with petroleum ether/dichloromethane (91:9, v:v); heavy aromatic hydrocarbon fraction with petroleum ether/dichloromethane (40:60, v:v); and resins with methanol/dichloromethane (65:35, v:v). Thin layer chromatography on Merck Kieselgel 60G (0.5 mm thick) was employed whenever the amount of maltenes available was either small (< 10 mg), or their content of light aromatic hydrocarbons was relatively high. The sample (~20 mg) was spotted 2 cm from the base of the plate and developed in *n*-hexane. *n*-Hexadecane, naphthalene, anthracene and pyrene were used as reference standards. The separated bands were visualised by spraying with a methanolic solution of Rhodamine 6G and viewing under UV light. Four fractions (saturated hydrocarbons; low-molecular-weight aromatic hydrocarbons; higher molecular-weight aromatic hydrocarbons; and polar compounds) were recovered from the silica gel by desorption using the same solvent mixtures as described for column chromatography. Finally, an aliquot (~10 mg) of the saturated hydrocarbon fraction was treated three times with urea to separate the normal and iso-alkanes (paraffins; adduct) from the cyclic and multibranched alkanes (naphthenes; non-adduct). Further details of this procedure may be found in Padley (1995).

2.3. Gas chromatography

The saturated hydrocarbon fraction (isolated from each of the bitumen samples attributed to data source 1 in Supplementary Table S1) was dissolved in *n*-hexane and analysed by gas chromatography (GC) in one of two laboratories, employing the following instrumental parameters.

In the Department of Geology and Geophysics, University of Adelaide, the instrument was a Varian 3400 gas chromatograph fitted with a DB-1 capillary column (30 m × 0.25 mm i.d. × 0.25 μm film thickness) and a flame ionisation detector (FID). The temperature program of the oven was 40 °C for 2 min, 40 to 300 °C at 4 °C/min and isothermal at 300 °C for 20 min. Two injection techniques were employed: a Varian split/splitless injector operated in the split mode (ratio 20:1) at a temperature of 300 °C, with hydrogen as the carrier gas at a head pressure of 15 psi; and a SGE on-column injector, with nitrogen as the coolant and hydrogen as the carrier at a linear flow rate of 35 cm/s.

At the Australian Geological Survey Organisation (now Geoscience Australia), Canberra, the instrument was a Hewlett Packard Series II 5890 gas chromatograph fitted with a FID and a HP Ultra-1 capillary column (50 m × 0.2 mm i.d. × 0.33 μm film thickness). The carrier gas was hydrogen at a linear flow rate of 35 cm/s. The sample was injected on-column at 60 °C and the oven temperature ramped from 60 to 300 °C at 4 °C/min before being held isothermal at 300 °C for 20 min.

All quantitative work was carried out in the former laboratory using the on-column injector and 3-methylheneicosane (AC 22; 10 μg/mL) as the internal standard.

2.4. High temperature capillary gas chromatography

High temperature capillary gas chromatography (HTCGC) of the total alkane fraction of six waxy bitumens (samples 28, 50, DB1, 48, 290 and EC1) was undertaken in the then Department of Environmental Sciences at the University of Plymouth, UK. Samples were dissolved in cyclohexane and heated to 55 °C prior to injection into a Carlo Erba HRGC 5300 Mega Series gas chromatograph fitted with a SGE HT5 column (12 m × 0.32 mm × 0.1 μm film thickness). The carrier gas was helium at 49 kPa head pressure and kept at a constant flow rate of 3 mL/min using the Carlo Erba CP-CF516 module. The sample was injected on-column with air as the coolant and the oven temperature-programmed from 50 to 410 °C at 10 °C/min before being held isothermal at 410 °C for 5 min. It should be noted that no attempt was made to quantify the recoveries of the saturated hydrocarbon fractions through the purification steps undertaken prior to their analysis by HTCGC (cf. Heath et al., 1995, 1997).

2.5. Gas chromatography–mass spectrometry

Selected total alkane and branched/cyclic (urea non-adduct) fractions were analysed by GC–MS in several modes: routine full scanning (SCAN), selected ion monitoring (SIM) and metastable reaction monitoring (MRM). However, only data from the MRM analyses are presented herein. Samples were dissolved in *n*-hexane and injected using an on-column injector at 50 °C with compressed air as the coolant. A magnetic sector double focussing VG 7070E mass spectrometer was utilised at the Australian Geological Survey Organisation, Canberra. The GC oven was programmed from 50 to 180 °C at 8 °C/min, 180 to 300 °C at 4 °C/min, and then held isothermal at 300 °C for 20 min. The capillary column was a HP Ultra-1 (50 m × 0.2 mm i.d. × 0.33 μm film thickness). The carrier gas was hydrogen at a linear flow rate of 30 cm/s. The operating conditions of the mass spectrometer were as follows: electron impact (EI) energy 70 eV, emission current 200 μA, source temperature 250 °C. Data were acquired and processed using VG 11-250 software. The concentrations of specific biological marker compounds were measured by the addition of a deuterated sterane standard 3-CD_3-5α(H)-cholestane (10 ng/mL). The biomarkers monitored included tricyclic terpanes, hopanes, diahopanes, methylhopanes, oleanane, bicadinanes, methylbicadinanes, steranes, diasteranes, and C_{30} desmethyl steranes and 4-methyl steranes, the derivation and environmental significance of which are summarised in Peters et al. (2005).

2.6. Stable carbon isotope mass spectrometry

Stable carbon isotopic compositions of the total saturated and aromatic hydrocarbon fractions were determined for the purpose of oil–oil correlations, as well as to give an indication of the bitumen's source affinity. Stable carbon isotopic analyses were carried out using a method slightly modified from that of Sofer (1984). Portions (~1.5 mg) of the saturated and aromatic hydrocarbon fractions were oxidised over cuprous oxide (CuO, 150 mg) at 900 °C for 8 h with silver wire (to remove sulfur and halogens) in evacuated quartz combustion tubes. The resultant CO_2 was separated from the co-produced H_2O and other condensable gases (e.g., SO_2) on a vacuum line by cooling with liquid nitrogen-chilled ethanol. The CO_2 was retrieved for introduction to the mass spectrometer by condensing the gas into collection tubes using liquid nitrogen. $^{13}C/^{12}C$ ratios were measured on a Micromass 602E isotope mass spectrometer in the Department of Geology and Geophysics, University of Adelaide. The instrument was equipped with a dual inlet system, allowing the sample to be compared directly with a reference gas. The resulting data are presented as $\delta^{13}C$ values relative to the VPDB standard. Reproducibility, as

determined from ten duplicate measurements of the TO_2 graphite standard, was ±0.06‰.

3. Results and discussion

3.1. Physical properties and bulk chemical composition

The suite of waxy coastal bitumens examined in this study comprises heavy to light, solid oils with densities varying between 10 and 40 °API (specific gravity = 0.9957–0.8270), pour points (43–82 °C) that reflect their high wax contents, and sulfur contents that range from low to moderate (0.1–2.6%) (Table 2). Collectively, they comprise inspissated residues of crude oils that range in composition from paraffinic, through paraffinic-naphthenic to aromatic intermediate (Fig. 2; based on data in Supplementary Table S2). Not surprisingly, waxy C_{23+} *n*-alkanes that commonly display an odd-over-even carbon number preference in the C_{25}–C_{31} range are prominent components of their saturated hydrocarbon fractions (Fig. 3). High-temperature gas chromatograms reveal the presence of individual *n*-alkanes up to C_{60+} (Fig. 4), implying that the parent crude oils are exceptionally rich in wax.

Most of these bitumens display evidence of pronounced weathering (as discussed in Section 3.3). Accordingly, each bitumen specimen has been assigned an empirical degree of weathering (DOW) value in the range 0–5, based upon the extent of removal of its normal and acyclic isoprenoid alkanes (Supplementary Tables S2 and S3).

3.2. Waxy bitumen families and their source affinities

Differences in the sulfur content, carbon isotopic composition and alkane biomarker geochemistry of the waxy bitumens permit their subdivision into five oil families. Based on a larger data set, the new classification (Table 3) expands the threefold genetic classification originally proposed by McKirdy (1984a, 1984b) and subsequently employed by McKirdy et al. (1986, 1988, 1994) to include two additional families, one marine (Family 4) and the other paludal (Family 5). The original classification invoked a similar suite of geochemical criteria, augmented by the hydrogen and sulfur isotopic compositions of the whole bitumen. Comparison of the new classification with those based on sample suites from the Northern Territory (Summons et al., 1993) and Western Australia (Alexander et al., 1994) reveals some overlap, but also significant differences (Table 3). Families 1 and 3 are common to all three classification schemes, whereas Families 2, 4 and 5 are not represented in either of these two earlier schemes.

The carbon isotopic signatures of the saturated and aromatic hydrocarbon fractions of the waxy bitumens (Supplementary Table S4) provide the first clue to their likely origin (Fig. 5). They reveal an algal source affinity for the vast majority of the specimens, while at the same time clearly distinguishing them from asphaltite, the other variety of southern Australian coastal bitumen (McKirdy et al., 1986, 1994, 2015; Volkman et al., 1992; Padley, 1995; Edwards et al., 1998, 2016; Hall et al., 2014). The lack of uniformity of the signatures plotted in Fig. 5 highlights the fact that, unlike the asphaltites, the waxy bitumens belong to multiple oil families.

Saturated hydrocarbon chromatograms typical of the five families are shown in Fig. 3. Of particular significance is the presence of botryococcane (peak labelled *), a C_{34} irregular acyclic isoprenoid alkane that elutes just before *n*-nonacosane (Peters et al., 2005), in 80% of the waxy bitumens examined. MRM chromatograms illustrating the distributions of triterpanes and steranes in Family 2 and 4 bitumens are shown in Figs. 6 and 7, while those of a Family 1 bitumen may be found in McKirdy et al. (1994, their Fig. 8).

Table 2
Summary of waxy bitumen bulk properties.[a]

Specific gravity, °API	Pour point[b], °C	Sulfur content, wt%	Oil type[c]
10–40	43–82	0.1–2.6	P (n = 16)
mean = 22	mean = 61	mean = 1.3	P-N (n = 36)
n = 42	n = 33	n = 45	Ar-I (n = 21)

[a] Based on information compiled in Supplementary Table S1.
[b] Measured using standard method IP 15/67.
[c] P = paraffinic, P-N = paraffinic-naphthenic, Ar-I = aromatic-intermediate (after Tissot and Welte, 1984).

The bicadinane and methylbicadinane distributions of Family 1 and 2 bitumens are compared in Fig. 8. A range of source-dependent biomarker signatures derived from 22 representative samples is presented in Tables 4a and 4b (parameters 1–16). For an explanation of the significance of these parameters the reader is referred to Peters et al. (2005) and references cited therein.

3.2.1. Family 1

The waxy bitumens of Family 1 are paraffinic to paraffinic-naphthenic crude oils with a relatively homogeneous bulk chemical composition and uniformly low sulfur contents (< 0.3%: Supplementary Tables S1 and S2; Fig. 2). Of all the samples analysed in this study they are the least depleted in ^{13}C (mean $\delta^{13}C_{sat} = -25.7$‰, mean $\delta^{13}C_{aro} = -24.5$‰: Supplementary Table S4; Fig. 5), likely due to the high content of *Botryococcus*-derived alginite in their source rocks (Boreham et al., 1994). This, in turn, is consistent with the mean isotopic composition of botryococcane ($\delta^{13}C = -12.4$‰), pristane (−25.9‰) and phytane (−26.6‰) in the seven Family 1 bitumens reported by Dowling et al. (1995).

The biomarker assemblage of the least weathered Family 1 waxy bitumen (sample 28, Fig. 3a) has the following pertinent characteristics (Tables 4 and 5): (i) a bimodal *n*-alkane distribution with maxima at C_{17} and C_{25}; (ii) an intermediate pristane/phytane ratio; (iii) a high concentration of botryococcane; (iv) an ethylcholestane-dominant 5α(H)-sterane distribution; (v) a diacholestane-dominant diasterane distribution; (vi) 4-methyl-24-ethylcholestane in abundance; (vii) the absence of dinosterane; (viii) a high C_{30} hopane/C_{29} 5α(H)-sterane ratio; (ix) the presence of 30-norhopane; (x) an abundance of oleanane, and (xi) relatively high concentrations of bicadinane and methylbicadinane.

The C_{15}–C_{35} *n*-alkane profile (Fig. 3a) is typical of oils derived either from a mixture of algal lipids (lower molecular weight *n*-alkanes) and terrestrial plant waxes (higher molecular weight *n*-alkanes), or from the freshwater chlorophyte alga *Botryococcus* sp.

With the advent of HTCGC it became possible to resolve and quantify high molecular weight (HMW) hydrocarbons in the range *n*-C_{40} to *n*-C_{100} (del Rio and Philp, 1992; Heath et al., 1995, 1997). High-temperature chromatograms of the saturated hydrocarbon fractions of five Family 1 waxy bitumens (Fig. 4a–e) reveal marked differences in their C_{35}–C_{70} *n*-alkane profiles. Making due allowance for the effect of biodegradation on the more volatile components of the chromatograms (discussed in Section 3.3), the differences are manifest in the relative proportions of two additional modes in their carbon number profiles (with maxima at C_{41}–C_{44} and ~C_{60}, respectively). Similar modes have been reported in the wax precipitated in pipes during production of Cenozoic lacustrine oil from the Redwash Field, Uinta Basin, Utah (Carlson et al., 1993). While the origin of this bimodal C_{35+} *n*-alkane distribution has yet to be established, the aforementioned differences likely point to variations in the mix of lacustrine algae and higher plants contributing resistant biopolymers (algaenans) and long-chain lipids to their respective source rocks (Tegelaar et al., 1989). Another feature reflecting the freshwater lacustrine source affinity of these Family 1 bitumens is the marked predominance

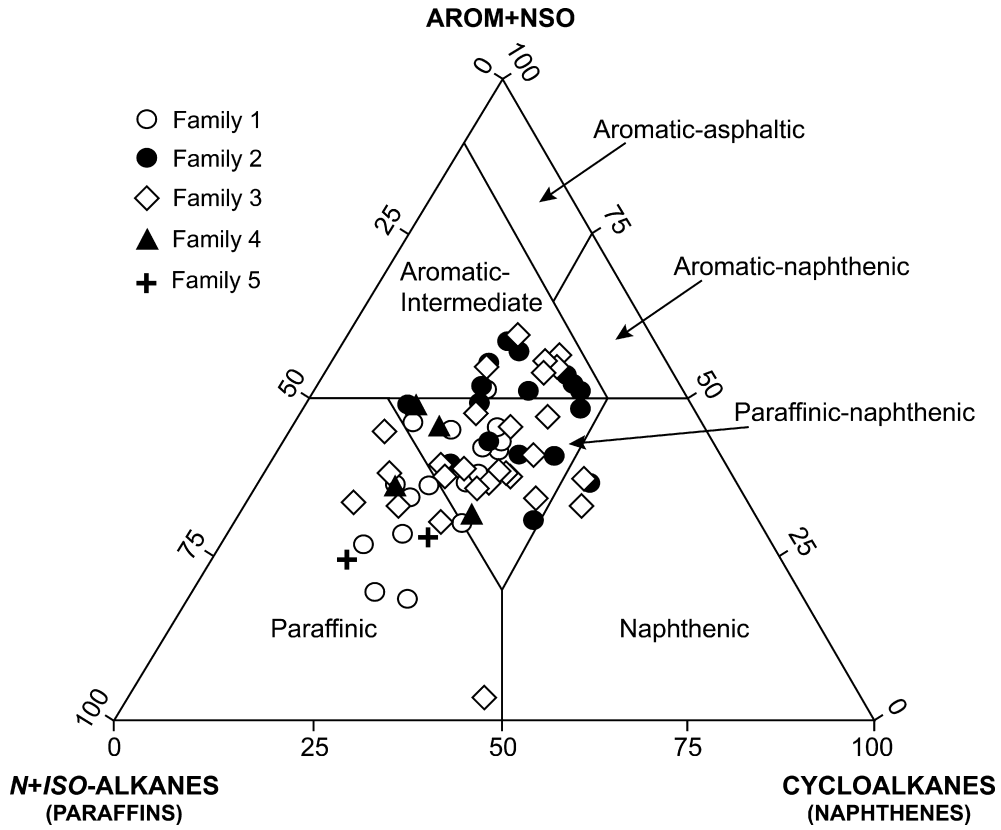

Fig. 2. Bulk composition of coastal waxy bitumens according to classification of Tissot and Welte (1984).

of normal over branched and monocyclic alkanes in the C_{40}–C_{60} range of their respective saturated hydrocarbon distributions (Fig. 4: Carlson et al., 1993).

Botryococcane is the saturated analogue of a C_{34} isoprenoid alkene synthesized by *Botryococcus braunii* (specifically clonal race B: Metzger et al., 1985). The presence of 4-methyl-24-ethylcholestane and the absence of dinosterane are together diagnostic of the input of freshwater dinoflagellates to their precursor organic matter (Summons et al., 1987). The pristane/phytane ratios of the least weathered Family 1 bitumens (Pr/Ph = <1–2.6: Supplementary Table S4) indicate that their precursor organic matter was deposited in an anoxic to sub-oxic lacustrine setting.

The sterane distribution of the Family 1 waxy bitumens is unusual in that the regular steranes are dominated by ethylcholestane, whereas diacholestane (not 24-ethyldiacholestane) is in most instances the dominant diasterane homologue (parameters 1 and 2, Table 4a). This anomaly does not appear to be the result of weathering because the sterane distribution is the same for the least and most weathered samples, and 24-ethyldiacholestane is not known to be preferentially removed by biodegradation (Connan, 1984; Peters et al., 2005).

The co-dominance of 24-ethylcholestane and diacholestane, therefore, is likely to be the result of the dual input of algal and land-plant detritus to the source kerogen. Another feature of the Family 1 waxy bitumens is their high abundance of 4-methyl-24-ethylcholestane relative to 24-ethylcholestane (C_{30} 4αMe-ster/C_{29} 5αH-ster = 2; parameter 12 in Table 3). Their lack of both 24-*n*-propylcholestane (a marine algal marker; Moldowan et al., 1990) and (in most instances) dinosterane confirms that the predominant algal contribution to their source material was from freshwater species.

The typically high abundance of diasteranes relative to steranes (parameter 5, Table 4a) in the Family 1 waxy bitumens reflects the high clay content of their source rock (McKirdy et al., 1983). Further evidence of an argillaceous source lithofacies comes from the dominance of the C_{30} homologue among the C_{27}–C_{35} hopanes and the presence of rearranged hopanes including C_{29}–C_{35} diahopanes and C_{27} and C_{29} 18αH-neohopanes (parameters 7–9, Table 4a; ten Haven et al., 1988; Moldowan et al., 1991).

In Family 1 waxy bitumens, the hopanes are dominated by αβ-hopane and the hopane/sterane ratio is high (C_{30} hop/C_{29} ster = 1.7–4.5; parameter 6, Table 4a) indicating a significant contribution of bacterial lipids to the parent organic matter. Such low sterane abundances relative to hopane are typical of lacustrine-sourced oils (Seifert and Moldowan, 1981). Methylhopanes are present in relatively low concentrations.

The angiosperm-derived pentacyclic triterpanes oleanane, bicadinane and methylbicadinane are abundant in the Family 1 waxy bitumens (Table 4b), indicating that they are derived from source rocks of Late Cretaceous or younger age (McKirdy et al., 1994 and References therein). Of all the waxy bitumens, those assigned to Family 1 have the highest concentrations of oleanane (ol/hop = 0.04–0.24; parameter 13, Table 4b). Methylbicadinane and four isomers of bicadinane were identified in the Family 1 waxy bitumens (Fig. 8). Using the nomenclature of Summons et al. (1993) and Murray et al. (1994a), the C_{30} bicadinane isomers are labelled W, T, T1 and R, respectively; and the methylated bicadinane isomers of T and T1, are annotated as MeT and MeT1, respectively. Typically, the relative abundance of bicadinane isomers in Family 1 waxy bitumens is T > T1 > R > W (parameter 16, Table 4b).

Other biomarkers that provide evidence for the contributions of higher plants to the precursor organic matter of the Family 1 waxy bitumens are diterpanes, notably 16βH-phyllocladane and the C_{18}–C_{20} labdanes (McKirdy et al., 1994). These diterpanes are derived from resin acids produced by both gymnosperms (mainly conifers) and angiosperms (Alexander et al., 1987).

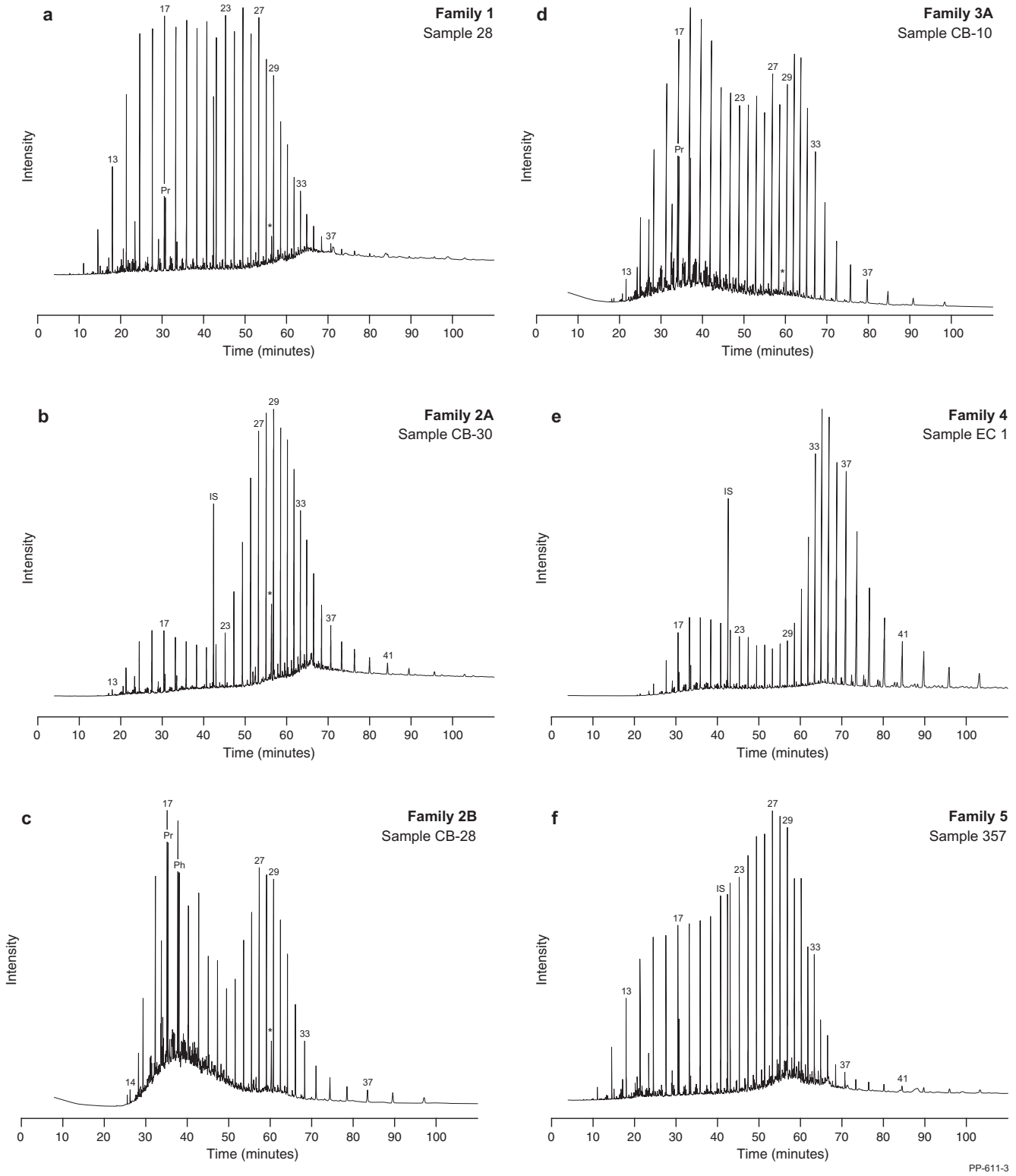

Fig. 3. GC-FID chromatograms of the saturated hydrocarbon fractions in representative waxy coastal bitumens. Key: Pr = pristane; Ph = phytane; * = C_{34} botryococcane; IS = internal standard, 3-methylheneicosane; numbers refer to chain length of *n*-alkanes.

Family 1 is an end-member group of high-wax, low-sulfur, paraffinic crude oils derived from an anoxic to suboxic lacustrine facies containing Type I sapropelic kerogen. In this instance, the precursors included freshwater algae (*Botryococcus* sp. and dinoflagellates) and the biodegraded remains of tropical land-plants (specifically angiosperms of the family *Dipterocarpaceae*). High concentrations of the following biomarkers are unique to the Family 1 waxy bitumens: botryococcane, 4-methyl-24-ethylcholestane, oleanane and bicadinane. *Botryococcus* sp. is common in non-marine kerogens but its diagnostic biomarker, botryococcane, is geologically rare. The limited occurrence of this saturated hydrocarbon may reflect both the rarity of *Botryococcus*

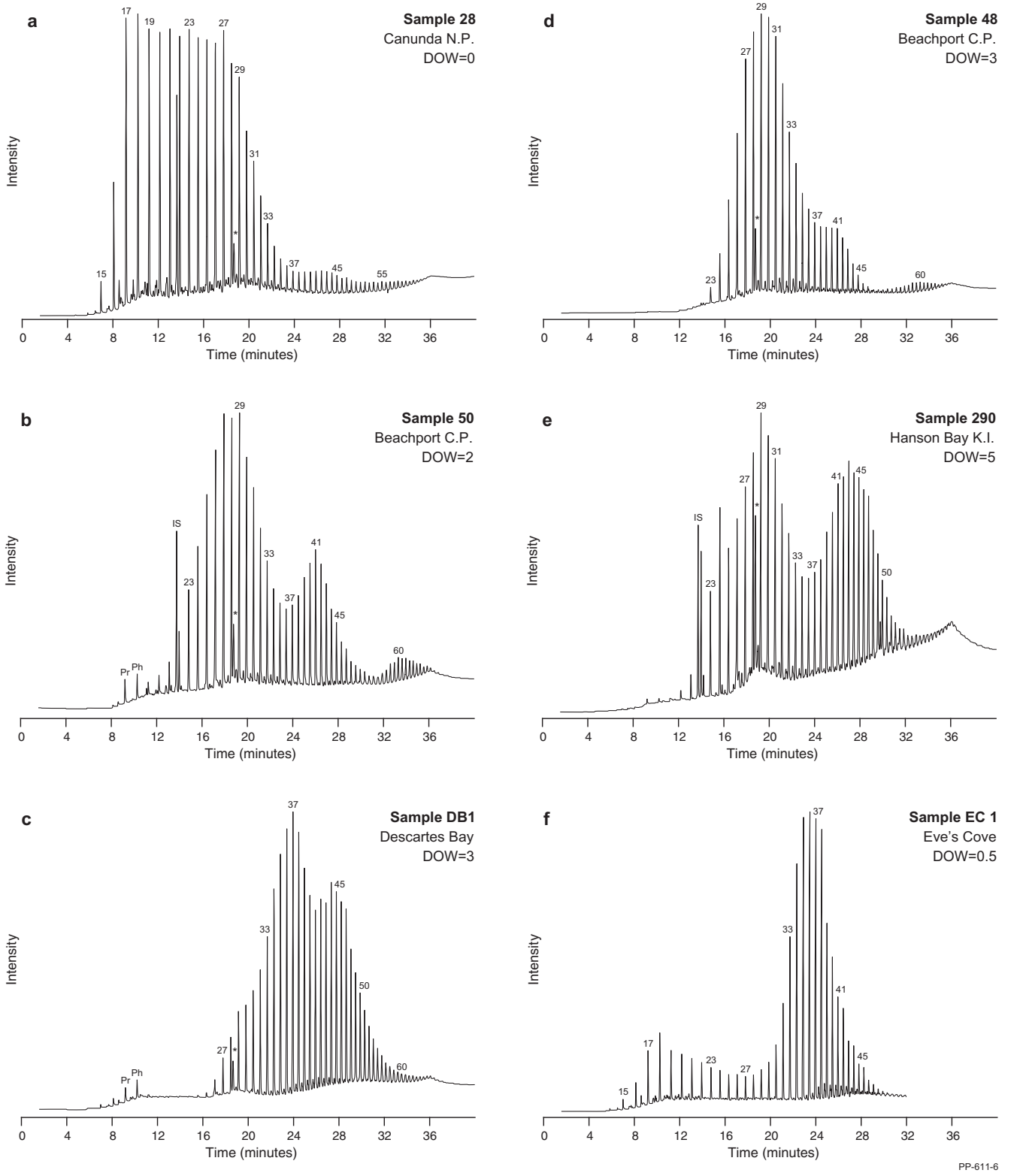

Fig. 4. High temperature GC-FID chromatograms of the saturated hydrocarbon fractions in representative waxy bitumens: Family 1 (a–e), showing effects of increasing weathering; Family 4 (f). Key as for Fig. 3.

braunii Race B (the only one of several different clonal races known to synthesize botryococcene) and the poor resistance of its biological precursor to bacterial and chemical degradation during deposition under oxic conditions (Derenne et al., 1988).

3.2.2. Family 2

The Family 2 waxy bitumens, like those of Family 1, also exhibit high concentrations of botryococcane (bot/*n*-C_{29} = 0.15–0.71; Table 5). However, their sulfur contents (0.6–2% S; Supplementary

Table 3
Comparison of oil family classifications for Australian waxy coastal bitumens.

Present study	Previous studies		Key biomarker characteristics	Source affinity
South Australia & western Victoria	Northern Territory	Western Australia		
Family 1	Family 1	Group 1	Botryococcane $C_{29} > C_{27} > C_{28}$ sterane C_{30} 4α-methylsterane Bicadinanes	Lacustrine shale Cenozoic
Family 2	NR	NR	As for Family 1, plus 24-*n*-propylcholestane & dinosterane	Lacustrine shale & marine carbonate Cenozoic
NR	NR	Groups 2 & 3	Bicadinanes abundant Oleanane/C_{30} hopane > 0.1	Deltaic Cenozoic
Family 3	Family 2	Group 4	C_{29}/C_{30} hopane > 1 C_{29} diasterane/sterane < 0.4 Bicadinanes & oleanane contents very low	Marine carbonate Cenozoic
NR	NR	Groups 5, 6 & 7	As for Group 4, but with significant oleanane & variable bicadinane contents	Marine carbonate Cenozoic
NR	NR	Group 8	$C_{27} > C_{29} > C_{28}$ sterane Bicadinanes & oleanane absent	Marine siliciclastic Mesozoic
Family 4	NR	NR	$C_{29} > C_{27} > C_{28}$ sterane C_{29}/C_{30} hopane > 1 Bicadinanes & oleanane absent	Marine carbonate & calcareous shale Mesozoic
Family 5	NR	NR	Pr/Ph ≥ 4 CV > 0.47	Non-marine paludal (coal)
Reference:	Summons et al. (1993)	Alexander et al. (1994)		

NR = not represented.

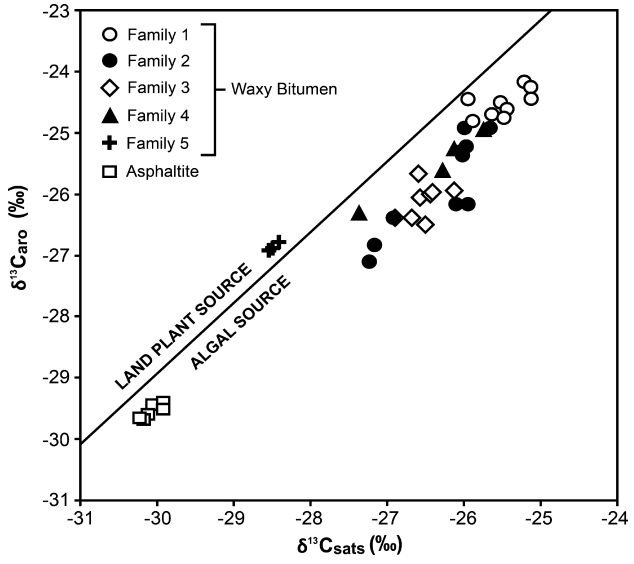

Fig. 5. Carbon isotopic compositions of C_{12+} saturated and aromatic hydrocarbon fractions in coastal waxy bitumens and asphaltites from southern Australia. The oblique line corresponds to a CV value of 0.47, where CV = −2.53 $\delta^{13}C_{sat}$ + 2.22 $\delta^{13}C_{aro}$ − 11.65, which according to Sofer (1984; reinterpreted by McKirdy et al., 1986) separates oils of land-plant origin from those of algal origin.

Table S1) are much higher than those of the Family 1 bitumens, and their carbon isotopic compositions are both lighter and far less uniform (Supplementary Table S4; Fig. 5). Their bulk chemical composition varies from paraffinic-naphthenic to aromatic-intermediate (Fig. 2) and the proportion of saturated to aromatic hydrocarbons also is highly variable (Supplementary Table S2). In the least weathered samples the Pr/Ph ratio is low (Pr/Ph = 0.7–1.7; Table 5). This second family of waxy bitumens may be subdivided into Family 2A and Family 2B, the latter distinguished by the presence of a pronounced naphthene 'hump' in their saturated hydrocarbon chromatograms (Fig. 3b and c). In the least weathered samples of both sub-families, the *n*-alkane profile is distinctly bimodal, although the mode centred on *n*-C_{17} is less dominant in Family 2A. It is largely on the basis of their bot/*n*-C_{29} values that little-weathered samples CB-28 and CB-92 (examples of Family 3 in McKirdy et al., 1994) are herein reassigned to Family 2B (Table 3).

The biomarker assemblage of this family is similar to that described for Family 1, except that in addition they contain trace quantities of the marine algal biomarkers, 24-*n*-propylcholestane and dinosterane (Tables 4 and 5). Their apparent mixed source affinity could arise in one of several ways: (i) generation of hydrocarbons from Type I/II to Type II/III kerogens with the organic matter being deposited in freshwater lacustrine or deltaic facies which were subjected to episodic marine incursions; (ii) acquisition of higher plant and/or marine signatures during migration of algal-derived lacustrine oils through deltaic and marine facies; (iii) reservoirs that have been charged by oils from different source rocks.

3.2.3. Family 3

The waxy bitumens of Family 3 differ from those of Family 2 in exhibiting a wider spread of bulk compositions (paraffinic to aromatic-intermediate; Fig. 2); lower concentrations of botryococcane (bot/*n*-C_{29} = 0.04–0.14; Supplementary Table S4); and, on average, higher sulfur contents (S = 1.1–2.6%; Supplementary Table S1) and somewhat lower pristane/phytane ratios (Pr/Ph typically < 1; Table 5). These latter features indicate that their source rocks were deposited beneath anoxic, saline (sulfate-bearing) bottom waters starved of iron. Their isotopic signatures overlap those of Family 2 waxy bitumens, but are much more tightly grouped (Fig. 5). As was the case for Family 2, these bitumens may be assigned to two sub-families, Family 3A and Family 3B, the saturated hydrocarbon chromatograms of the latter displaying a more pronounced naphthene 'hump' than that shown in Fig. 3d. The presence of this feature suggests that for both Family 2B and Family 3B bitumens, the reservoir of the parent oil received multiple

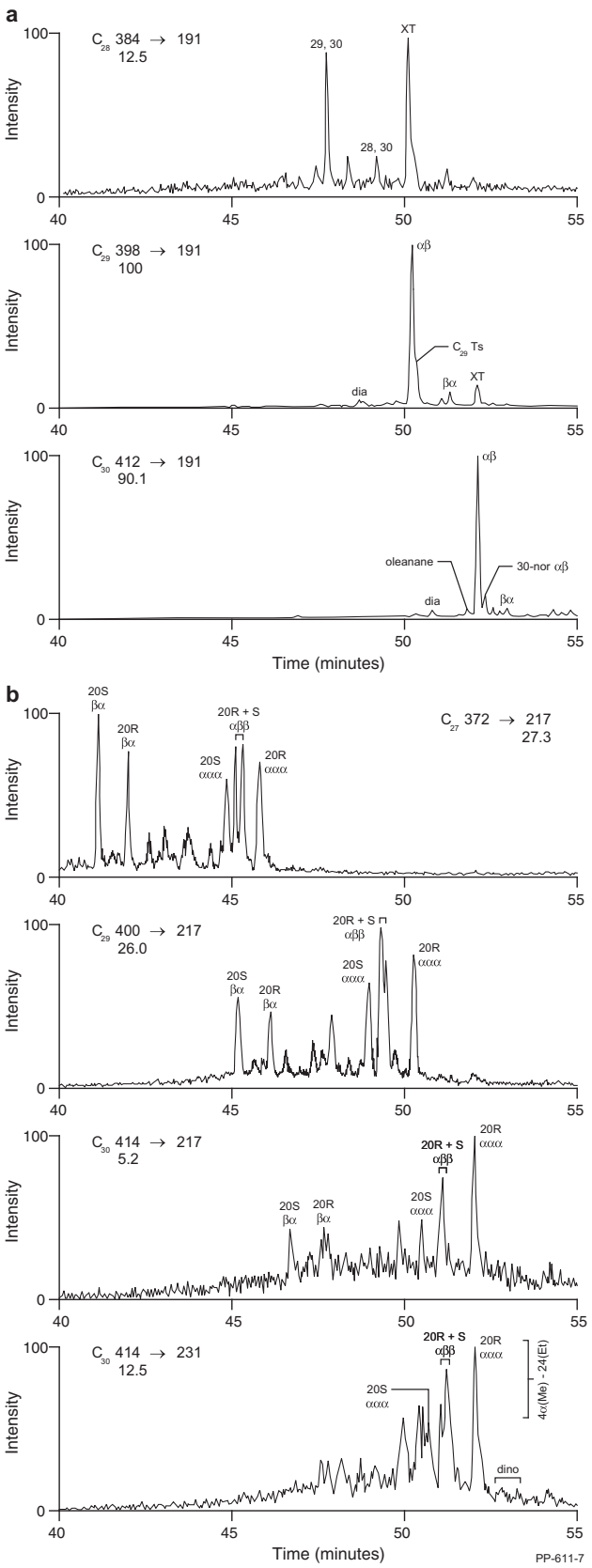

Fig. 6. MRM chromatograms showing the distribution of (a) triterpanes ($M^+ \rightarrow m/z$ 191) and (b) steranes ($M^+ \rightarrow m/z$ 217 and 231) in a Family 2 waxy bitumen (sample CB-28).

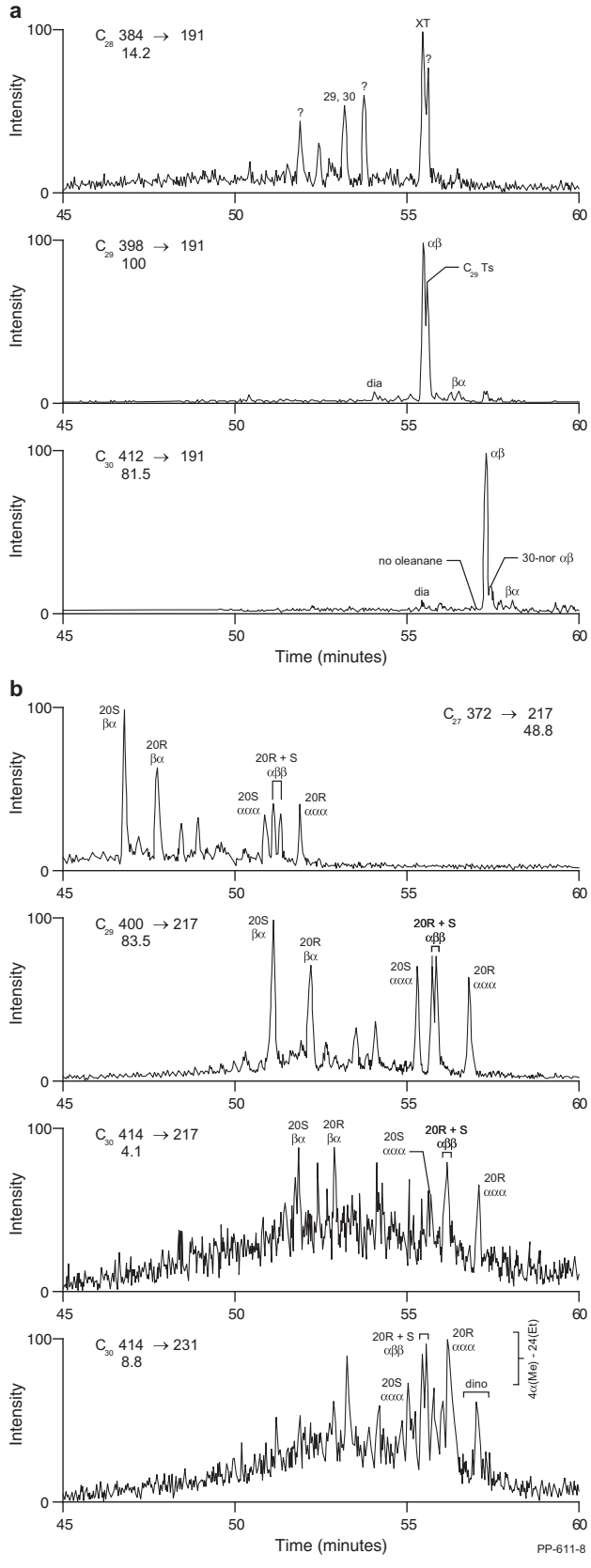

Fig. 7. MRM chromatograms showing the distribution of (a) triterpanes ($M^+ \rightarrow m/z$ 191) and (b) steranes ($M^+ \rightarrow m/z$ 217 and 231) in a Family 4 waxy bitumen (sample CB-31-2).

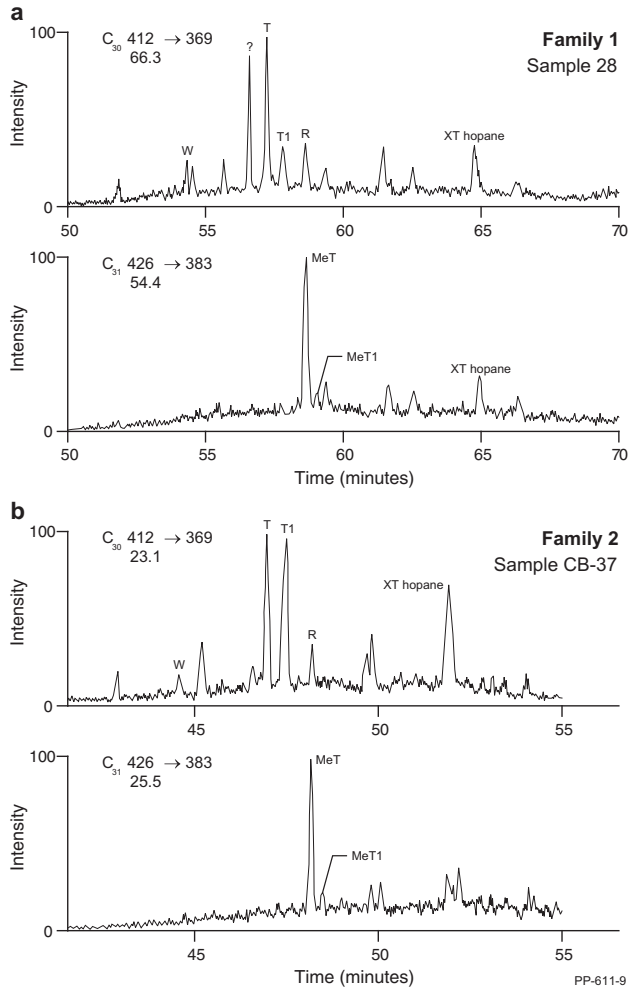

Fig. 8. MRM chromatograms showing the contrasting bicadinane and methylbicadinane signatures of representative Family 1 and Family 2 waxy bitumens.

charges, of which an earlier one underwent in situ biodegradation before migration into the trap resumed.

The Family 3 waxy bitumens can be further distinguished from those of Families 1 and 2 by the dominance of cholestane and diacholestane in their C_{27}–C_{30} 5α(H)-sterane and diasterane distributions, the presence of 24-*n*-propylcholestane and dinosterane, and the fact that bicadinane and oleanane are present in only trace quantities (Tables 4 and 5). The Family 3 parent oils were generated from a mixture of algal lipids and higher plant waxes, with the former providing the greater input to their source kerogen. The change in overall geochemical character from Family 1 to 3 reflects a progressive decrease in the amount of terrestrial influence and a corresponding increase in marine algal source affinity. An accompanying decline in the triterpane ratio Ts/Tm, from values ≥ 3 in Family 1 to ≈ 1 in Family 3 (Table 4a), is consistent with a change in source lithofacies from anoxic shale for Family 1 to predominantly carbonate for Family 3 (Peters et al., 2005).

3.2.4. Family 4

Family 4 comprises seven miscellaneous samples of waxy, paraffinic to paraffinic-naphthenic bitumen (Fig. 2) which have bimodal *n*-alkane profiles, with maxima at C_{18} and either C_{31} or C_{34}, but notably lack botryococcane. For such a little-weathered bitumen (DOW = 0.5), the saturated hydrocarbon distribution of sample EC 1 (Figs. 3e and 4f) is highly unusual in that it is dominated by *n*-alkanes in the range C_{30}–C_{50}. It bears a striking resemblance to that of wax precipitated from a marine-sourced crude oil during production (Philp, 1994, his Fig. 2).

MRM data were acquired for four of these samples (Tables 4a and 4b). The presence of trace quantities of 24-*n*-propylcholestane and dinosterane, but no oleanane or bicadinanes (Fig. 7) provides evidence for their marine source affinity. In the absence of appropriate MRM data showing its lack of bicadinanes, sample CB-31-1 was originally assigned to Family 2 (McKirdy et al., 1994). The relatively low abundance of diasteranes in sample EC 1 (C_{29} dia/ster = 0.18), together with the obvious presence of 30-norhopanes (C_{29}/C_{30} hopane = 1.8) and its bulk C-isotopic composition (CV < 0.47; Fig. 5), implies that its parent oil was derived from a marine carbonate source rock containing predominantly algal organic matter. Higher diasterane abundances in the other three samples are more typical of an argillaceous source rock. This implied difference in source facies in turn is consistent with the fact that sample EC1 is appreciably more depleted in ^{13}C than the other Family 4 bitumens and hence plots as an outlier in Fig. 5. It is therefore apparent that the Family 4 waxy bitumens originated from several different source rocks of varying marine lithofacies. The highly weathered state of more than half the samples assigned to this family (Supplementary Table S3) precluded their definitive assignment to sub-families.

The waxy bitumens of Family 4 differ from those of Family 1, 2 and 3 in lacking botryococcane, oleanane and bicadinanes (Table 3). While not necessarily precluding an origin in Southeast Asia, the absence of these biomarkers raises the possibility that the Family 4 waxy bitumens could be products of a petroleum system located within the offshore Otway Basin and/or Bight Basin, although the Bremer Basin and offshore Perth Basin could also be possibilities given the west to east current direction that transports the waxy bitumens in southern Australian waters (Edwards et al., 2016). Further afield, the basins on Australia's North West Shelf are also worthy of consideration since they lie in the path of the Leeuwin Current.

3.2.5. Family 5

Family 5 comprises five samples of soft bitumen colonised by goose barnacles (Edwards et al., 2016, their Fig. 5g). These waxy bitumens have a paraffinic bulk composition (Fig. 2) and are sulfur-poor (S = 0.2%: Supplementary Table S1). Their *n*-alkane distribution is unimodal (maximum at C_{27}) with the lower molecular weight homologues (C_{15}–C_{21}) largely intact (Fig. 3f). Pristane is much more abundant than phytane (Pr/Ph > 4). The other key characteristics which distinguish the Family 5 waxy bitumens from Families 1 to 4 are that they do not contain botryococcane, they are isotopically more depleted in ^{13}C, and their CV values are >0.47 (Supplementary Table A4), highlighting their land-plant source affinity (Fig. 5). Together these features indicate their likely derivation from coal-rich fluvial to deltaic sediments. MRM analysis of their saturated hydrocarbon fractions is required to further constrain the age and location of their source lithofacies.

3.3. Weathering characteristics

The weathering of an oil at or near the sea surface involves a combination of evaporation, photo-oxidation, water washing and biodegradation and causes a marked change in its chemical composition (National Research Council, 2003). The most significant effects are evaporative loss of the light ends (C_1–C_{12}), dissolution of monoaromatic hydrocarbons (e.g., benzene, toluene) and the selective removal of the C_{12}–C_{23} *n*-alkanes by bacteria, followed at a slower rate by the low molecular weight isoprenoid alkanes. Overall, there is an increase in the proportion of non-hydrocarbons (resins and asphaltenes), mainly at the expense

Table 4a
Source-dependent biomarker parameters of selected waxy bitumens.

Sample ID	Steranes					Triterpanes			
	C_{27}:C_{28}:C_{29} ster	C_{27}:C_{28}:C_{29} dia	27/29 ster	30/29 ster	29 dia/ster	30/29 hop/ster	27 Ts/Tm	29/30 hop	30 dia/hop
Family 1									
28	37:26:37	51:21:28	1.0	0	0.70	2.8	3.7	0.61	0.17
4066	21:25:54	nd	0.38	0	0.70	3.5	3.6	0.84	0.19
Gelt (318)	22:20:58	45:22:33	0.38	0	0.44	3.1	2.1	0.82	0.17
Devil's (317)	29:22:49	35:31:33	0.57	0	0.20	1.7	0.77	1.0	0.09
CB-38 (378)	32:19:49	11:37:52	0.65	0	0.46	4.2	2.7	0.68	0.10
59	nd	nd	0.68	0	0.41	4.5	3.1	0.51	0.09
A923.152A	x	x	x	x	x	x	3.2	1.0	0
48 Dune	x	x	x	x	x	x	3.0	0.88	0.21
148 Dune	28:21:51	39:26:34	0.56	0	0.65	2.1	3.1	0.90	0.21
68	x	x	x	x	x	x	2.7	0.49	0.18
290	30:18:52	45:21:34	0.59	0	0.46	2.9	3.5	0.78	0.11
Family 2									
CB-37	35:11:54	nd	0.64	0.22	0.60	6.7	2.3	0.52	0.11
CB-92 (606)	30:20:50	45:25:30	0.61	0.22	0.22	1.4	1.1	1.3	0
CB-28 (605)	37:26:37	42:28:30	0.98	0.23	0.32	1.1	1.4	1.2	0.03
Family 3									
CH 8	41:22:37	37:32:31	1.1	0	0.26	0.61	1.3	1.5	0
CB-35	43:25:32	47:24:29	1.3	0	0.32	1.3	1.0	1.2	nd
CB-8	43:19:38	37:23:40	1.1	0.05	0.25	0.93	1.1	1.7	0.04
Family 4									
CB31-1 (379)	33:14:53	47:22:31	0.62	nd	0.60	0.78	3.3	1.2	nd
CB31-2 (608)	37:20:43	49:16:35	0.87	0.05	0.96	0.97	3.4	1.0	nd
EC 1	nd	nd	0.83	nd	0.18	nd	0.63	1.8	0
ST 15	nd	nd	nd	nd	0.35	1.8	1.9	2.1	0
Parameter	1	2	3	4	5	6	7	8	9

Ratios calculated from peak areas in MRM mass chromatograms as follows:
1. C_{27}:C_{28}:C_{29} 5α,14α,17α (H) (20*R*) steranes (M^+ 372: 386: 400 → *m/z* 217); 2. C_{27}:C_{28}:C_{29} 13β,17α (H) (20*R*) diasteranes (M^+ 372: 386: 400 → *m/z* 217); 3. C_{27} 5α,14α,17α (H) cholestane (20*R*)/C_{29} 5α,14α,17α (H) ethylcholestane (20*R*) (M^+ 372 → *m/z* 217) and (M^+ 400 → *m/z* 217); 4. C_{30} 5α,14α,17α (H) *n*-propylcholestane (20*R*)/C_{29} 5α,14α,17α (H) ethylcholestane (20*R*) (M^+ 414 → *m/z* 217) and (M^+ 400 → *m/z* 217); 5. C_{29} 13β,17α (H) diasteranes (20*S* + 20*R*)/C_{29} 5α,14α,17α (H) steranes (20*S* + 20*R*) (M^+ 400 → *m/z* 217); 6. C_{30} 17α,21β (H)-hopane/C_{29} 5α,14α,17α (H) steranes (20*S* + 20*R*) (M^+ 412 → *m/z* 191) and (M^+ 400 → *m/z* 217); 7. C_{27} 18α (H)-22,29,30-trisnorneohopane/C_{27} 17α (H)-22,29,30-trisnorhopane (M^+ 370 → *m/z* 191); 8. C_{29} 17α,21β (H)-hopane/C_{30} 17α,21β (H) hopane (M^+ 398 → *m/z* 191) and (M^+ 412 → *m/z* 191); 9. C_{30} 17α (H) diahopane/C_{30} 17α,21β (H)-hopane (M^+ 412 → *m/z* 191).

of the saturated hydrocarbons as illustrated by sub-sampling of Family 1 bitumen sample 28 (Supplementary Table S2).

The aromatic hydrocarbon fractions of 11 samples (representative of all but Family 4) were analysed by SIM GC–MS to augment the limited aromatic maturity data already available on southern Australian waxy bitumens (McKirdy et al., 1994). However, inspection of their respective mass chromatograms revealed that in more than half the samples phenanthrene and the methylphenanthrenes, along with naphthalene and the methylnaphthalenes, had largely been removed by weathering. A combination of water washing and biodegradation (Palmer, 1993; Peters et al., 2005) is likely to have been responsible for their removal.

The Family 1 waxy bitumens include some of the most highly weathered samples (i.e., *n*-alkanes < C_{23} and light isoprenoid alkanes totally removed). These paraffinic to paraffinic-naphthenic bitumens typically contain high concentrations of saturated hydrocarbons (Supplementary Table S2) which would not normally be expected to remain in extremely weathered oil samples. A selection of these bitumens with DOW values in the range 0–5 exhibit a progressive depletion of the C_{14}–C_{32} *n*-alkanes (Fig. 4a–e) which is typical of oils that have been degraded by aerobic bacteria (Bailey et al., 1973). The Pr/Ph ratio systematically decreases with increased weathering as pristane is more volatile than phytane. A marked increase in the pristane/*n*-heptadecane and the phytane/*n*-octadecane ratios is observed as biodegradation proceeds (Supplementary Table S4). The botryococcane/*n*-nonacosane ratio in both Family 1 and Family 2 bitumens likewise increases with increasing biodegradation.

In some of the Family 1 waxy bitumens analysed by MRM (A923.152A, 48 Dune and 68), the sterane distributions have been impacted by weathering (DOW = 2–4; Supplementary Table S2), displaying a preferential loss of the biological isomer (20R). Two of these samples have also lost their bicadinanes and methylbicadinanes (parameter 16, Table 4b). That said, there are other similarly weathered samples in which these biomarkers are intact, although this may also reflect variations in the input of dipterocarp resins to the source rocks.

The waxy bitumens of both Family 2B and 3B exhibit a pronounced naphthene 'hump' in their saturated hydrocarbon chromatograms (e.g., Fig. 3c), and yet in some samples their mid-range (C_{13}–C_{17}) alkanes remain. The 'hump' therefore may be an inherited characteristic resulting from multiple phases of oil emplacement, and at least one episode of in-reservoir biodegradation, prior to seepage. The extensive biodegradation displayed by the most severely weathered waxy bitumens is most likely to have occurred during their exposure to the marine environment. However, another possibility is that their parent oils may have been degraded in the reservoir. For example, similar highly biodegraded oils are produced from the Duri Field in Sumatra (Seifert and Moldowan, 1981). From the biomarker information obtained for these waxy bitumen samples it is not possible to definitively differentiate between in situ reservoir alteration and biodegradation in the marine environment.

The fact that the waxy bitumens form tar balls means that they can survive in the ocean for long (albeit unquantified) periods of time. Their resistance to weathering is not unique as there are

Table 4b
Source-dependent biomarker parameters of selected waxy bitumens.

Sample ID	C_{30} Steranes			Triterpanes			
	24-*n*-propyl	dino	4 α-Me/29 ster	ol/hop	bicad T/hop	ol/∑bicad	Major bicadinanes
Family 1							
28	A	A	nd	0.09	0.66	0.09	T, MeT, R, T1, W
4066	A	A	nd	0.04	nd	nd	T, MeT, T1, R, W
Gelt (318)	A	A	nd	0.08	0.37	nd	T, T1, R, W*
Devil's (317)	A	A	nd	0.24	1.5	nd	T, MeT, T1, R, W
314	A	A	nd	nd	2.9	nd	T, T1, MeT, W
CB-38 (378)	A	A	2.1	0.08	0.48	0.09	T, MeT, T1, R, W
59	A	P?	nd	0.05	nd	nd	x
A923.152A	A	A	nd	nd	2.1	0.31	T, MeT, T1, R, W
48 Dune	A	A	nd	0.06	nd	nd	x
148 Dune	A	A	nd	0.04	1.2	0.03	T, T1, R, W*
68	A	A	nd	0.09	nd	nd	T, MeT, T1, R, W
290	A	P?	nd	0.07	0.46	0.10	T, MeT, T1, R, W
Family 2							
CB-37 (604)	P	P	5.5	0.06	0.22	0.24	T, T1, MeT, R, W
CB-92 (606)	P	P	0.88	0.05	nd	nd	T1, MeT, T, R, W
CB-28 (605)	P	P	0.61	0.05	0.07	0.61	T, MeT, T1, R, W
Family 3							
CH8	nd	nd	nd	x	0.14	x	T, MeT, T1, R, W
CB-35	A	A	0.83	nd	0.08	nd	T1, T, MeT, R, W
V2-6	P	P	nd	x	x	x	x
CB-8	P	P	0.28	0.02	nd	nd	T1, T, R, W*
Family 4							
CB-31-1 (379)	P	P	0.18	0	0	0	A
CB-31-2 (608)	P	P	nd	0	0	0	A
EC 1	nd	nd	nd	0	0	0	A
ST 15	nd	nd	nd	0	0	0	A
Parameter	10	11	12	13	14	15	16

Ratios calculated from peak areas in MRM mass chromatograms as follows:
10. 24-*n*-propylcholestane (M^+ 414 → *m/z* 217; 11. dinosterane (4,23,24-trimethylcholestane) (M^+ 414 → *m/z* 231); 12. 4α-methyl-24-ethylcholestane (20*R*) + 4α-methyl-23,24-dimethylcholestane (20*R*)/24-ethylcholestane (20*R*) (M^+ 414 → *m/z* 231); 13. oleanane/C_{30} 17α,21β (H) hopane (M^+ 412 → *m/z* 191). 14. bicadinane T/C_{30} 17α,21β (H) hopane (M^+ 412 → *m/z* 369) and (M^+ 412 → *m/z* 191); 15. oleanane/bicadinanes T + T1 + R (see Fig. 8) (M^+ 412 → *m/z* 191) and (M^+ 412 → *m/z* 369); 16. Bicadinanes listed in order of decreasing abundance. Isomers labelled as in Grantham et al. (1983), Alam and Pearson (1990), Summons et al. (1993) and McKirdy et al. (1994). * Methylbicadinane M^+ 426 → *m/z* 383 not recorded. A = absent; P = present; nd = not determined because peak not integrated or appropriate MRM chromatogram not acquired; x = not determined because compound(s) removed by severe weathering.

Table 5
Diagnostic characteristics and source affinity of waxy bitumen families.

Family	S wt%	Pr/Ph	BI[a]	Bot/*n*-C_{29}	Steranes	24-*n*-Propyl	Dino
1 n = 35	< 0.3	0.2–2.6	530–870	0.16–0.86	29 > 27 > 28	A	A
2 n = 17	0.6–2	0.7–1.7	nd	0.15–0.71	29 ≥ 27 > 28	P	P
3 n = 33	1.1–2.6	0.5–1.1	104–173	0.04–0.14	27 > 29 > 28	P or A	P or A
4 n = 7	0.9	0.5–1.4	0	0	29 ≥ 27 > 28	P	P
5 n = 5	0.2	4–11	nd	nd	nd	nd	nd

	Hop/Ster	Oleanane	Bicadinanes	CV[b]	Source affinity
1	1.7–4.5	P	P	< 0.47	Lacustrine shale Anoxic–suboxic Cenozoic
2	1.1–6.7	P	P	< 0.47	Lacustrine shale > marine carbonate Anoxic–suboxic Cenozoic
3	0.6–1.3	P	P	< 0.47	Marine carbonate > lacustrine shale Anoxic/euxinic Cenozoic
4	0.8–1.8	A	A	< 0.47	Marine carbonate or calcareous shale Anoxic–suboxic Mesozoic
5	nd	nd	nd	> 0.47	Non-marine (paludal) coal Oxic

Pr/Ph = pristane/phytane; Bot = C_{34} botryococcane.
[a] Botryococcane Index = C_{34} botryococcane × 100/$\sum C_{37} - C_{40}$ head-to-head isoprenoid alkanes (Seifert and Moldowan, 1981).
[b] CV = −2.53 $\delta^{13}C_{sat}$ + 2.22 $\delta^{13}C_{aro}$ − 11.65 (Sofer, 1984). See Table 3 for key to other parameters.

many documented cases of large pieces of bitumen surviving in the sea for more than a year (Blumer et al., 1973; Butler et al., 1976; Lee, 1980; Warnock et al., 2015).

3.4. Evidence for a Southeast Asian origin

The unusual assemblage of the age-specific biomarkers botryococcane, oleanane and bicadinanes found in the Family 1, 2 and 3 waxy bitumens (Table 5), suggests that their parent oils originated from one or more Cenozoic basins somewhere in the Indonesian Archipelago (Fig. 9).

Botryococcane has previously been identified only in Cenozoic lacustrine oils and source rocks from Central Sumatra (notably the Minas, Duri and South Bekasap fields; Seifert and Moldowan, 1981) and the Eocene Maoming oil shale, Maonan Basin, China (Brassell et al., 1986, 1988). High concentrations of 4-methyl-24-ethylcholestane are also diagnostic of lacustrine environments. This sterane is abundant in the Messel Shale (Rubinstein and Albrecht, 1975) and many non-marine Chinese oils (Shi et al., 1982; Wolff et al., 1986a, 1986b; Philp and Fan, 1987; Fan and Zhang, 1988). However, the association of high concentrations of botryococcane and 4-methylsteranes has been rarely documented, a notable exception being the aforementioned Maoming oil shale. Oleanane first appeared during the Late Cretaceous (late Campanian), but high concentrations of this pentacyclic triterpane have only been observed in Cenozoic sediments and oils, corresponding to the time when angiosperms became the dominant flora. Bicadinane has been identified in Cenozoic oils throughout Indonesia, southern China, Papua New Guinea, the Philippines and New Zealand (van Aarssen et al., 1990; Murray et al., 1993, 1994a, 1994b). However, high concentrations of bicadinane appear to be confined to Cenozoic sediments and associated oils from Southeast Asia.

Bicadinanes are derived principally from tropical angiosperms of the family *Dipterocarpaceae*, trees that first appeared in the Eocene and now dominate the lowland rain forests of Southeast Asia, but do not grow in Australia (Paijmans, 1976; Ashton, 1982; Ghazoul, 2016). Fossil evidence suggests that they originated in Gondwana, and subsequently migrated from India to Asia (Dutta et al., 2011). However, no dipterocarp micro- or megafossils have been identified in Cenozoic sedimentary successions within Australia (Hill, 1994). The trace levels of bicadinanes (± oleanane) found in a Jurassic crude oil and source rock from the Eromanga Basin (Armanios et al., 1995), Jurassic oils and condensates from the Perth Basin (Summons et al., 1995) and Late Cretaceous-sourced oils in the Gippsland Basin (George et al., 1998; Volk et al., 2011) are attributable to a more general angiosperm flora (van Aarssen et al., 1994; Murray et al., 1994b). Therefore, it seems reasonable to conclude that these Family 1–3 waxy bitumens do not originate from Australian sedimentary basins.

High concentrations of bicadinane are in fact typical of Cenozoic oils and source rocks from Indonesia (Cox et al., 1986; Teerman et al., 1987; van Aarssen et al., 1992; Sosrowidjojo et al., 1994) and Papua New Guinea (Murray et al., 1993; Waples and Wulff, 1996). They reflect the predominance of dipterocarps in the Cenozoic flora of this region. Given that the biomarker signatures of the complete range of waxy bitumens indicate their derivation from lacustrine through marginal marine to paludal (coal-bearing) organic facies (Tables 3 and 5), it is informative to note similar signatures and source affinities in oils from Indonesia (Table 6).

Waxy oils of lacustrine source affinity are confined to basins located in western Indonesia (Fig. 9). Arguably the best documented are those in the onshore Central and South Sumatra basins and the offshore Sunda and East Java basins (Pramono et al., 1990; Schiefelbein and Cameron, 1997; Hwang et al., 2002). Their respective source rocks, viz. the Brown Shale Formation, Pematang Group (Williams et al., 1985) and the Banuwati Shale (Robinson, 1987), are of Eocene to Oligocene age. The former unit generated the oil reserves of the giant Minas (4 billion barrels) and Duri (3 billion barrels) fields. Lacustrine oils likewise occur further north in the offshore East Natuna, West Natuna and Anambas basins (Noble et al., 2009).

Seifert and Moldowan (1981) were the first to report that lacustrine oils produced from the Central Sumatra Basin have divergent geochemical signatures. The biomarker assemblage of the heavily biodegraded Duri-1 crude oil matches that of the Minas-1 oil, both containing abundant botryococcane. In other oils, such as the South Bekasap-1 crude, the concentration of botryococcane is much lower; and in the Pematang-1 oil this freshwater algal marker is entirely absent, a more saline lagoonal setting for its source rock being indicated by the presence of gammacerane (Table 6). With access to larger datasets, subsequent workers have divided the Indonesian lacustrine oil family into subgroups: Type A, B and C (ten Haven and Schiefelbein, 1995) and Groups I–V (Hwang et al., 2002). Across Sumatra this compositional heterogeneity has been attributed to differences in water chemistry in different sub-basins (Katz and Mertani, 1989) or, alternatively, variation of the Brown Shale source rock facies within a single deep lake system (Schiefelbein and Cameron, 1997).

The biomarker assemblages of the southern Australian waxy bitumens (particularly Families 1–3) show many similarities to some of the lacustrine western Indonesian oils (Table 6), but an exact match is not evident. For example, the deep lacustrine Minas-1 oil from the Central Sumatra Basin is a high wax, low sulfur, paraffinic crude oil that closely resembles the Family 1 bitumens. As previously reported (Seifert and Moldowan, 1981; Currie et al., 1992; Summons et al., 1993; McKirdy et al., 1994; Peters et al., 2005), its biomarker signatures include unusually high concentrations of botryococcane, C_{30} 4-methylsteranes, bicadinane and methylbicadinane; a ethylcholestane-dominant sterane distribution; the presence of oleanane; and an absence of C_{30} 24-*n*-propylcholestane and dinosterane, all features of the Family 1 bitumens (Table 5). Moreover, its C-isotopic signatures ($\delta^{13}C_{sat} = -26.2‰$, $\delta^{13}C_{aro} = -24.7‰$, CV = −0.17; Peters et al., 2005) also plot close to those of the Family 1 bitumens (Fig. 5). However, its bicadinane distribution differs significantly from that of a typical Family 1 bitumen (Fig. 10), as does its HMW C_{40}–C_{60} alkane distribution and those of several other fresh/brackish water lacustrine oils from Central Sumatra (Carlson et al., 1993), preventing their exact correlation. On the other hand, the Family 1 bicadinane distribution does resemble those of oils from the Ardjuna Sub-basin, Java, and the Lufa district of Papua New Guinea.

Other Indonesian waxy crude oils originated from coal-rich fluvial deltaic sediments of Oligocene-Miocene age and are assigned to the terrigenous family (Table 6). Source rocks in this category include the coals of the Sihapas Group, Central Sumatra Basin and the coals and shales of the Talang Akar Formation in the South Sumatra Basin, the Sunda Basin and the Ardjuna Sub-basin (Robinson, 1987; Pramono et al., 1990; Noble et al., 1991). The Australian waxy bitumens of Family 5 (Table 5) are insufficiently characterised to ascertain the degree to which they resemble the terrigenous oils of western Indonesia, but warrant further research.

With the exception of several examples from the North Sumatra and East Natuna basins, most Indonesian crude oils of marine origin are located in the eastern half of the archipelago (ten Haven and Schiefelbein, 1995; Noble et al., 2009). Here their source rocks may be of either Cenozoic or Mesozoic age and include both carbonate and shale lithofacies, whereas in western Indonesia oil-prone marine source rocks are both less common and all of Cenozoic age (Price et al., 1987; Robinson, 1987; Peters et al., 1999). It is in Eastern Indonesia that most of the known offshore oil seeps occur (Fig. 9). Those located in Seram and the Timor Sea are of marine source affinity and thought to have originated in Mesozoic car-

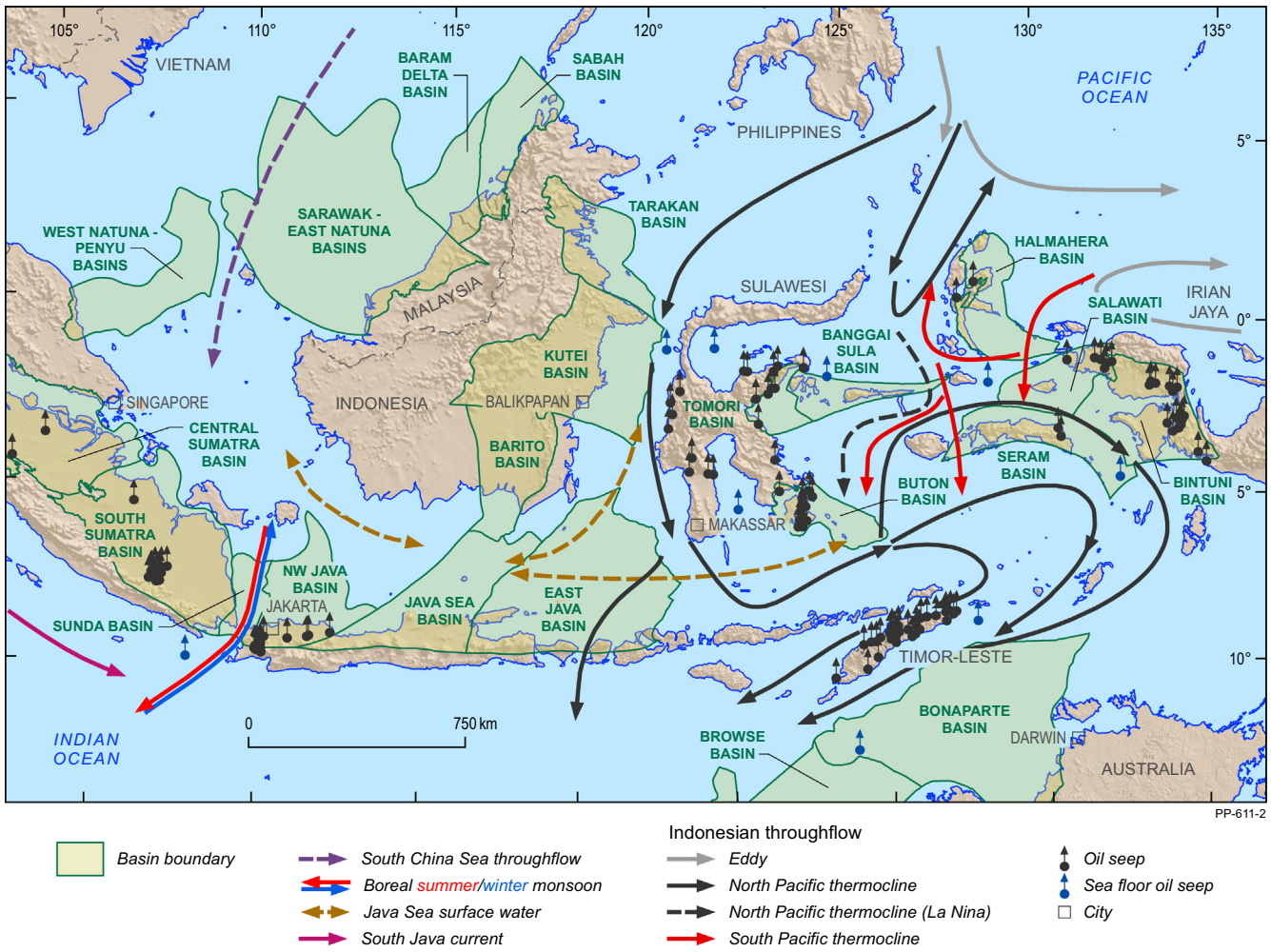

Fig. 9. Map of selected sedimentary basins within the Indonesian Archipelago and environs showing the location of oil seeps (after Livsey et al., 1992; Macgregor, 1995; Noble et al., 2009) and pathways of the Indonesian and South China Sea throughflows (after Gordon, 2005; Gordon et al., 2012; Susanto et al., 2016). Sedimentary basin outlines are from Robertson (2017).

bonate and shale lithofacies (Price et al., 1987; Noble et al., 2009). In this respect, they appear similar to the Family 4 waxy bitumens which notably lack oleanane and bicadinanes (Table 5).

An alternative origin for the marine waxy bitumens of Family 4 is from the Australian North West Shelf and Perth basins where marine oils are known (Edwards and Zumberge, 2005 and References therein). In contrast to Southeast Asia, the passive margin of Australia has few documented seepage sites. Where detected, seepage appears to be predominantly gas rather than oil (Logan et al., 2010). However, in the Browse Basin on the Yampi Shelf (in the vicinity of the Cornea field) natural oil and gas seeps have been sampled (Rollet et al., 2006; Burns et al., 2010), and a micro-seep of oil was also found at one site in the southern Bonaparte Basin (Stalvies et al., 2017; Fig. 9). These seeps were correlated with crude oils (Cornea and Gwydion) from the Yampi Shelf and determined to be sourced by the Early Cretaceous Echuca Shoals Formation (Logan et al., 2008; Stalvies et al., 2017). However, these oils (e.g., Cornea-1: $\delta^{13}C_{sat} = -28.6‰$, $\delta^{13}C_{aro} = -27.7‰$, CV = −0.8 2) and source rocks (Edwards and Zumberge, 2005) are more depleted in ^{13}C than are the Family 4 waxy bitumens (Fig. 5), ruling out the Browse Basin as their potential source. Likewise, the Early Triassic-sourced marine oils from the Perth Basin have significantly more depleted isotopic compositions than the Family 4 waxy bitumens and contain a distinctive C_{33} *n*-alkylcyclohexane (Summons et al., 1995; Thomas and Barber, 2004) not found in the bitumens, thereby precluding their correlation. The majority of Jurassic marine oils in the Northern Carnarvon and Bonaparte basins are non-waxy crudes sourced from organic-rich marine shales within the Dingo Claystone and Vulcan Formation (Edwards and Zumberge, 2005). However, since these basins also host some waxy Jurassic oils that are isotopically similar to the Family 4 waxy bitumens, further work is required to evaluate their possible common origin.

In summary, the southern Australian coastal waxy bitumens cannot yet be precisely correlated with any Southeast Asian oils. Nevertheless, the Family 1–3 waxy bitumens (comprising ~80% of the samples examined in this study) display sufficient similarities with oils from Indonesia to be most likely derived from this region. An Australian origin for the Family 4 and 5 waxy bitumens remains a possibility, notwithstanding the fact that they do not match any of the oils so far discovered in the Otway Basin, nor any of the minor oil shows (e.g., Greenly-1, Duntroon Sub-basin; Jerboa-1, Eyre Sub-basin) encountered during drilling in the Bight Basin (Padley, 1995; Edwards et al., 1999; Ruble et al., 2001). A potential origin for the Family 5 bitumens in the Gippsland Basin was considered but dismissed because, although Latrobe Group-sourced oils have similar pristane/phytane ratios (Pr/Ph = 3–11), they are significantly less depleted in ^{13}C ($\delta^{13}C_{sat} = -25.2$ to $-28.0‰$; $\delta^{13}C_{arom} = -24.6$ to $-26.9‰$; Summons et al., 2002). Further studies of potential source rocks and generated hydrocarbons within the basins along Australia's southern margin

Table 6
Biomarker characteristics of western Indonesian oil families.

Family	Key biomarker signatures	Basins	Source lithofacies & age	Data sources
Fresh/deep lacustrine	Botryococcane C_{30} Tetracyclic polyprenoid (TPP)[a] C_{30} 4-methylsteranes C_{26}/C_{25} tricyclic terpane $\gg 1$ Ts/Tm > 1 Oleanane Bicadinanes Hopanes > Steranes Pristane/phytane = 1.5–3 24-*n*-Propylcholestane absent	Central Sumatra South Sumatra Sunda	Shale Cenozoic	1, 2, 3, 4, 7
Brackish lacustrine	As above, but with minor or no botryococcane & higher abundances of terrigenous triterpanes (oleanane, bicadinanes) Gammacerane present	Central Sumatra South Sumatra Ombilin East Java	Shale Cenozoic	1, 2, 3, 4, 7
Terrigenous (fluvio-deltaic)	C_{29} sterane dominant Bicadinanes Oleanane C_{26}/C_{25} tricyclic terpane < 1 Pristane/phytane > 3	North Sumatra Central Sumatra South Sumatra West Natuna Northwest Java	Coal, carbonaceous shale Cenozoic	2, 3, 4, 5
Marine	C_{27} sterane dominant 24-*n*-Propylcholestane present C_{29}/C_{30} hopane > 1 Pristane/phytane = 1–2.5	North Sumatra East Natuna	Carbonate Cenozoic	2, 3, 4, 6

Key to data sources: 1. Seifert and Moldowan (1981); 2. Robinson (1987); 3. ten Haven and Schiefelbein (1995); 4. Schiefelbein and Cameron (1997); 5. van Aarssen et al. (1992); 6. Subroto et al. (1992); 7. Hwang et al. (2002).
[a] Reported as unknown 'lacustrine classifier' by ten Haven and Schiefelbein (1995); subsequently identified by Holba et al. (2000).

and North West Shelf are required for definitive oil-source and oil-oil correlations.

3.5. Possible modes of origin

Accepting that the majority of the waxy coastal bitumens are the remains of weathered Southeast Asian crude oils, their occurrence in southern Australia has several possible explanations: jettisoned oil cargoes, tanker washings, leakage from oil pipelines, or natural ocean flotsam emanating from oil seeps.

Australia began importing crude oil during the mid-1950s and continues to import large quantities from Southeast Asia. The composition of many of these imported crudes is documented in a national oil-on-the-sea database (NOSID; Sandison and Edwards, 2001). Oil slicks from tanker washings and petroleum accidentally spilled from local shipping intermittently strand along the Victorian and South Australian coastline (Padley et al., 1993). The physical and chemical properties of these freshly spilled oils readily distinguish them from weathered waxy bitumen (Edwards et al., 2016). The latter is therefore thought not to be a by-product of local maritime activity in southern Australian waters. However, the Straits of Malacca and the Sunda Strait are major shipping routes where tanker cleaning operations are undertaken prior to reloading at Singapore (Fig. 9). Regular oil spills in the western Java Sea since 2004 have been linked to both the high density of maritime traffic and leakage from submarine pipelines carrying oil from offshore production platforms (Putri et al., 2016).

Active onshore oil seeps are common throughout the Indonesian Archipelago (Fig. 9) and Papua New Guinea (Livsey et al., 1992; Waples and Wulff, 1996), but few overlie known oil fields and none of the giant fields are associated with seeps (Macgregor, 1993, 1995). In the case of the giant Minas Field, the low ratio of gas to oil has reduced the capacity for vertical migration, making it difficult for the oil to escape to the surface (Thompson et al., 1991). Unless adjacent to major rivers, the inland locations of these surface seeps make them less likely sources for the Australian waxy bitumens.

Submarine seeps such as those recently identified by multibeam echo sounder bathymetry and backscatter imaging in combination with deep-sea piston coring (Fig. 9: Noble et al., 2009; Orange et al., 2009) are potential sources for the Australian coastal waxy bitumens. As previously mentioned, the seeps located offshore from the islands of Timor and Seram are potential points of origin for the marine Family 4 waxy bitumens. It is also worth noting that the asphalt deposit on Buton (Fig. 9) originates from a marine crude oil (Satyana et al., 2013). While no seeps have yet been identified in the offshore Sunda Basin (Macgregor, 1995), this Cenozoic lacustrine depocentre is the site of major lacustrine oil production and remains a possible source for the Family 1 bitumens. Likewise, the co-occurrence of Cenozoic lacustrine and marine oils in the offshore East Natuna Basin (Noble et al., 2009) makes it a potential source for the mixed lacustrine/marine bitumens of Families 2 and 3 (Table 5). Natural seafloor seepage of crude oil into the ocean throughout the Indonesian Archipelago is now well established and would readily give rise to the seaborne waxy bitumens (tar balls) that ultimately strand in southern Australia. However, the fact that such seeps are generally episodic and ephemeral (Kvenvolden and Cooper, 2003) adds to the difficulty of linking a given bitumen stranding to its parent seep.

The possibility that Southeast Asian oils could be transported as flotsam into Australian waters via the Leeuwin Current (McGowran et al., 1997) was invoked by Currie et al. (1992) as an explanation for the geochemical similarity of the Western Australian coastal bitumens and certain Indonesian crude oils. For details of the complex system of ocean currents that flow through the Indonesian Archipelago (Fig. 9) the reader is referred to Gordon (2005), Gordon et al. (2012) and Susanto et al. (2016), while the manner in which they feed into the headwaters of the Leeuwin Current and ultimately into the coastal current and South Australian Current that traverse Australia's southern continental margin (Middleton and Bye, 2007) is summarised in Edwards et al. (2016). A new study, aimed at more closely linking the stranding pattern of coastal bitumen in South Australia to offshore oceanographic conditions during the last three years (2014–2016), is well advanced (Corrick et al., 2016). Meanwhile, the data presented

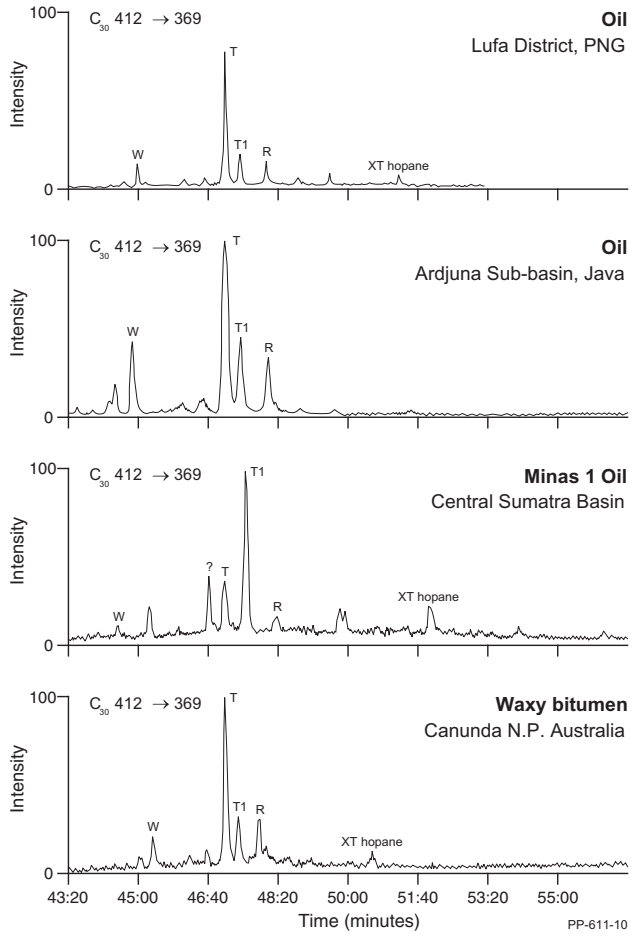

Fig. 10. Partial MRM M^+ 412 → *m/z* 369 chromatograms illustrating the C_{30} bicadinane distributions in a Family 1 waxy bitumen and crude oils from Indonesia and Papua New Guinea.

herein show that ~80% of the waxy bitumens stranded on the ocean beaches of South Australia and western Victoria between 1960 and 1992 did not originate in Australia, and are not derived from either exploration activity or natural seepage in the adjacent Bight or Otway basins.

4. Conclusions

Geochemical analysis of 96 waxy bitumens recovered from the coastline of southern Australia has shown them to be inspissated high-wax crude oils of paraffinic to aromatic-intermediate bulk composition. Subtle elemental, isotopic and biomarker differences evident in these bitumens allow their assignment to at least five different oil families. Their inferred source facies range from deep freshwater lacustrine through paludal to estuarine and deltaic marine, possibly deposited within different sedimentary basins.

The Family 1, 2 and 3 waxy bitumens all contain biomarkers derived from the freshwater alga *Botryococcus* sp. and tropical angiosperms (notably dipterocarps). Similar biomarker assemblages are unknown in Australian crude oils and source rocks but are common in the Cenozoic basins of Indonesia. The Family 4 bitumens lack these biomarkers, but do contain dinosterane and 24-*n*-propylcholestane, indicative of a marine source affinity, while the C-isotopic signatures and high Pr/Ph ratios of the Family 5 waxy bitumens are consistent with an origin from coal-rich source rocks. Hence, it is believed that the majority of the waxy bitumens represent an oceanic influx of non-indigenous, Indonesian crude oil into the waters of southern Australia. Further studies on the Family 4 and 5 waxy bitumens are required, since their origin from seepage in offshore Australian basins has not been ruled out.

The body of geochemical and oceanographic evidence assembled herein points to the origin of the Family 1–3 waxy bitumens from long-term natural seepage within the Indonesian Archipelago. However, it is unknown whether their parent oils emanate from submarine seeps or, less likely, from inland seepages which are then carried to the sea by rivers. The common practice of tanker cleaning operations in the Java and Banda seas may augment the supply of coastal bitumen to the beaches of Australia. Further work is required to determine whether such an anthropogenic input makes a significant contribution to the natural background level of seaborne oil within the region.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at https://doi.org/10.1016/j.orggeochem.2017.12.010.

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